The coherence scale of correlated Kondo impurities

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The coherence scale of heavy fermion systems is the onset temperature of the Kondo singlet formation. We show by studying the two-impurity Anderson model that this scale strongly deviates from the single-ion Kondo temperature in the presence of Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. It scales as the RKKY interaction or the single-impurity Kondo temperature when either of them becomes dominant. This can be understood as a two-stage Kondo process, the formation of a partially screened Kondo state at the coherence scale, and the formation of coherence (local) Fermi liquids at another lower energy scale. The latter is due to the competition between a fully screened Kondo state and the inter-impurity spin singlet state. Relevance of these energy scales to heavy fermion systems is also discussed.

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Recently there has been much debate on the origin of the coherence scale T_{coh} for heavy fermion systems, which characterizes the evolution of the local or itinerant nature of the strongly interacting f-electrons [1, 2]. This scale is manifested in various thermodynamics, transport and magnetic responses measurements, for instance, the entropy $S(T) = R\ln 2$ for $T \geq T_{coh}$. Traditionally, it is associated with the single-ion Kondo temperature T_K^0, below which, localized f-electrons form a Kondo-singlet resonance state with conduction electrons at the chemical potential and becomes itinerant to form a heavy Fermi liquid band. However, analysis on a group of heavy fermion materials exhibiting quantum critical behaviors reveals that it is rather associated with the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction I, which is the spin exchange interaction between magnetic ions [1]. A solution to this discrepancy relies on the understanding of the effect of RKKY interaction on the Kondo singlet formation. While the associated lattice models for heavy fermion systems are difficult to solve, the two-impurity Anderson or Kondo model provides a minimal setting to study such a competition effect. An antiferromagnetic (AFM) RKKY interaction ($I > 0$) favors an inter-impurity spin singlet state between local moments against the formation of Kondo singlet state. Previous studies have been focused on the associated quantum phase transition or crossover between these two distinct states [3–11]. In the particle-hole symmetry case, a low energy scale is uniformly suppressed to zero when the ratio I/T_K^0 is tuned to a critical value, indicating a continuous phase transition [3]. Without particle-hole symmetry, a parity splitting term is found to smear out the sharp transition behavior [10, 11]. However, the characterization for all the energy scales present in this system, and therefore a complete interpretation for the transition are still lacking.

In this Letter, we adopt the numerical renormalization group (NRG) method [12] to study the two-impurity Anderson model, with a focus on identifying and characterizing all the energy scales from the $T = 0$ dynamical quantities, such as the single-particle Green's function, uniform and staggered spin susceptibilities. We show results on two systems: one with the fixed T_K^0 and tunable I and another for two impurities sitting on nearest neighbor sites of a real lattice. In the latter system, the generated I is positive and the ratio I/T_K^0 is tuned by the hybridization between the local orbitals and conduction electrons. We find that a two-stage Kondo process, through a partially screened Kondo state, is very important in understanding the phase transition. Accordingly, there are in general two energy scales present, the coherence scale T_{coh} as the crossover from the high temperature free moment state with the impurity entropy $S = 2\ln 2$ to the partially screened Kondo state with $S = \ln 2$, and a lower energy scale T_{FL} as onset of coherent Fermi liquid behaviors. In the first system, T_{FL} is associated with the energy gap T^* between the inter-impurity spin singlet state and the triplet state, which is also the onset for either the full Kondo resonance or the inter-impurity spin singlet state where $S = 0$ for both states. In the second system, a parity splitting term T_m, which generates Kondo resonance at the chemical potential, is present and determines T_{FL} as it is in general bigger than T^* when $I \sim T_K^0$. However, T_{coh} has the same behavior in both systems: it is determined by either I or T_K^0 when whichever is dominant while a value higher than both when $I \sim T_K^0$. We argue that this high energy scale persists in the lattice model. It indicates that in quantum critical metals, where $I > T_K^0$ is suggested, I serves as the coherence scale, or the onset scale for the formation of partial Kondo screening. The magnetic phase transition relies on additional competing effects involving the scales T^* and T_m.

The Hamiltonian for the two-impurity Anderson model can be written as

$$\begin{aligned} H &= \sum_{\mathbf{k}\sigma} \varepsilon_{\mathbf{k}} c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} + \frac{1}{\sqrt{N}} \sum_{\mathbf{k}\sigma i} (V_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}_i} c^\dagger_{\mathbf{k}\sigma} f_{i\sigma} + \text{h.c.}) \\ &+ \sum_{i\sigma} \varepsilon_f f^\dagger_{i\sigma} f_{i\sigma} + \sum_i U n_{fi\uparrow} n_{fi\downarrow} \,, \end{aligned} \tag{1}$$

where $i = 1, 2$. This describes two interacting local orbitals $f_{i\sigma}$ (Anderson impurities) in hybridization with a non-interacting conduction electron band $c_{\mathbf{k}\sigma}$ with the strength $V_{\mathbf{k}}$ at each impurity site $\mathbf{r}_i$. The local orbitals have the local energy level ε_f and onsite Coulomb interaction U. In the single occupancy limit, this model can be mapped into a two-impurity Kondo model by a canonical transformation [11]. It is convenient to take linear combinations of the local orbitals in terms of even or odd parities with respect to their center

$f_{(e,o)\sigma} = (f_{1\sigma} \pm f_{2\sigma})/\sqrt{2}$ while the fluctuations due to conduction electrons can be casted into two channels with the hybridization functions $\Gamma_{p\sigma}(\omega)$,

$$\Gamma_{p=(e,o)}(\omega) \;=\; -\frac{1}{2}\mathrm{Im}\left[\frac{1}{N}\sum_{\mathbf{k}}\frac{V_{\mathbf{k}}^2|e^{i\mathbf{k}\cdot\mathbf{r}_1} \pm e^{i\mathbf{k}\cdot\mathbf{r}_2}|^2}{\omega - \varepsilon_{\mathbf{k}} + i0^+}\right]. \quad (2)$$

This problem is amenable to the numerical renormalization group method, which iteratively solves the problem by gradually approaching low energies in logarithmic scale. From the NRG spectrum, we can calculate the dynamical quantities, $G_{AB}(\omega) = -i\int_0^\infty dt e^{i\omega t}\langle [A(t), B(0)]_\pm\rangle$, in particular, the Green's function $G_{fp\sigma}$ with $A = B^\dagger = f_{p\sigma}$, the uniform and staggered spin susceptibilities, with $A = B = (S_{1z} + S_{2z})/\sqrt{2}$ and $(S_{1z} - S_{2z})/\sqrt{2}$, respectively. For these quantities, we adopt the recently developed complete-Fock-space method [13], which conserves the total spectral weight and has better accuracy in the intermediate and high energy range. Details of our calculation will be presented elsewhere [14].

We consider two systems of interest, isolated impurities (with $|\mathbf{r}_1 - \mathbf{r}_2|$ being infinite), and impurities sitting on nearest neighbors (that is, separated by the lattice constant a), respectively. We assume $V_{\mathbf{k}} = V$ and a three-dimensional tight-binding dispersion of conduction electrons $\varepsilon_{\mathbf{k}} = -2t\sum_{i=1}^{3}\cos k_i a$. We set $6t = 1$ as the energy unit. For these two cases, the hybridization functions can be well represented by $\Gamma_{e,o}(\omega) = \Gamma_0$ and $\Gamma_{e,o} = \Gamma_0(1 \mp \omega)$ near the chemical potential, where $\Gamma_0 \sim \rho_0 V^2$ and ρ_0 is the conduction electron density of state. For simplicity, we adopt these forms for the whole band in our calculation.

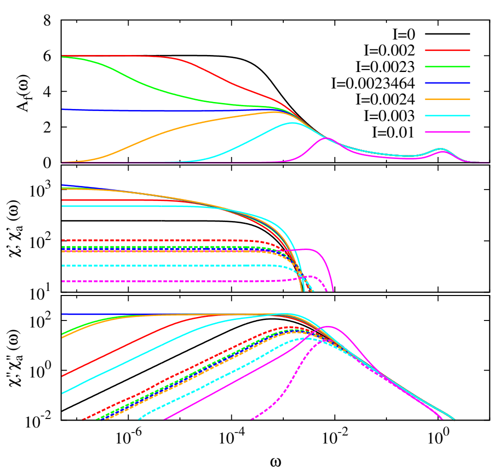

FIG. 1: (color online) Spectral functions (A_f), real and imaginary parts of the spin susceptibilities as functions of the energy for various values of the RKKY interaction in the isolated impurities case. Both uniform (χ) and staggered (χ_a) are shown in the same figure, and represented by dotted and solid lines, respectively. Here $\Gamma_e = \Gamma_o = 0.045\pi$ and particle-hole symmetry is assumed with $U = 2$ and $\varepsilon_f = -U/2$. While only the positive energy range is shown, quantities in the negative energy range are symmetric or antisymmetric accordingly. The spectral function is also symmetric for different parities and spins in this case.

In Fig. 1, we show the results of the spectral function $A_f(\omega) = -\mathrm{Im}G_f(\omega)/\pi$, uniform ($\chi$) and staggered ($\chi_a$) spin susceptibilities for the isolated impurities case. We find that $\chi = \chi_a$ (curves labeled with $I = 0$), which indicates the two impurities indeed appear as two separate Kondo scatterers without any correlation $\langle S_{1z}S_{2z}\rangle = 0$. Below the orbital level $|\varepsilon_f|$ or $\varepsilon_f + U$, there is only one energy scale present in all these quantities, i.e., the single-ion Kondo temperature, below which $A(\omega) \approx 1/(\pi\Gamma_0)$ as the Kondo resonance and the spin susceptibilities take the (local) Fermi liquid forms. From $\chi'(0) = 1/(4T_K^0)$, we determine $T_K^0 = 1.02 \times 10^{-3}$, which is consistent with the energy where dynamical quantities change behaviors. To turn on the correlations, we include an explicit RKKY term $I\mathbf{S}_1 \cdot \mathbf{S}_2$. Upon increasing I for $I > 0$, the spectral function firstly increases to about half weight at an energy scale T_K^* around T_K^0, then either increases again to the full weight or decreases to 0 at a lower energy scale T^*. T^* is uniformly suppressed to 0 at $I_c \approx 2.3T_K^0$, which is consistent with the two-impurity quantum critical point obtained earlier [3]. In the uniform spin susceptibility, only the high energy scale T_K^* is present, this is manifested as the peak position in the imaginary part, under which $\chi''(\omega) \sim \omega$; also as $\chi'(0) = 1/(4T_K^*)$. In the staggered spin susceptibility, both energy scales are present. $\chi''_a \sim \omega$ when $\omega < T^*$ while $\chi''_a \sim const$ when $T^* < \omega < T_K^*$. The real part $\chi'_a(0) \sim 1/T^*$, which becomes divergent as $T^* \to 0$. Two energy scales are also present in thermodynamics calculations [10, 15], where the impurity entropy evolves from $2\ln 2 \to \ln 2 \to 0$ at T_K^* and T^*, respectively.

The two-energy-scale scenario signifies a two-stage Kondo process [16], i.e., as energy (or temperature) is lowered, the system has to go through an intermediate state existing within $T^* < \omega < T_K^*$, which is also the quantum critical state for $T^* = 0$. From the analysis on the NRG spectrum, also suggested by the conformal field theory [6] and Bosonization method [7, 9], this state is governed by fluctuations between two almost degenerate states, the spin singlet configuration of two local orbitals and one of the triplet configurations, $|\uparrow\uparrow\rangle + |\downarrow\downarrow\rangle$. These two states form a doublet and can be represented by local fermion operators d and $d^\dagger$, with $d^\dagger d = 0$, 1 accordingly. In the Bosonization approach[9], it is found that only one combination (or one Majorana fermion) couples to the extended fermions, $(\psi_{sf}(0) - \psi_{sf}^\dagger(0))(d + d^\dagger)$, where ψ_{sf} is the spin-flavor degree of freedom. This indicates that only half of the impurity spins is Kondo-screened with the entropy $\ln 2$. The Majorana fermion is also responsible for the divergent staggered spin susceptibility and non-Fermi liquid behaviors. [We also notice that the Green's function of d is consistent with our numerical calculation that $-\mathrm{Im}\Sigma \approx \Gamma_0$ (not shown) and $A_f \approx 1/(2\pi\Gamma_0)$.] A finite $(I_c - I)d^\dagger d$ acts as the gap between the doublet which suppresses this type of excitations. Depending on whether the triplet or singlet configurations become the ground state, the system either becomes a full Kondo resonance state (Kondo singlets) or a pseudo-gap (inter-impurity spin singlet) state to further reduce the entropy. Both states are Fermi liquid fixed points with the scattering phase shifts (for each channel) $\delta = \pi/2$ or 0 while the leading irrelevant operators lead to Fermi liquid behaviors. Therefore, T^* is the characteristic scale for this gap and acts as the (coherent) Fermi liquid temperature T_{FL}. Though T^* is in fact a pseudo-gap scale in inter-impurity spin singlet phase, we find that χ" and χ_a" are linear in ω, and $A_f \sim \omega^2$, which distinguish this phase from the free spin state.

In another aspect, we can associate T_K^* as the coherence scale T_{coh}, because it indeed indicates the onset of Kondo singlet formation, though to a partially screened state. As I increases, T_{coh} becomes slightly bigger than T_K^0 (in comparison, T_{coh} decreases from T_K^0 when $I<0$) and eventually scales as I when $I \gg T_K^0$, e.g, $I = 0.01$ in Fig. 1. In the latter limit, while the inter-impurity spin singlet is the ground state which doesn't favor Kondo coupling, at the energy scale I, excitations to spin triplet become possible while the Kondo resonance emerges. This can also be evidenced from the spin susceptibility, which has a Pauli term superposed on the Van Vleck form.

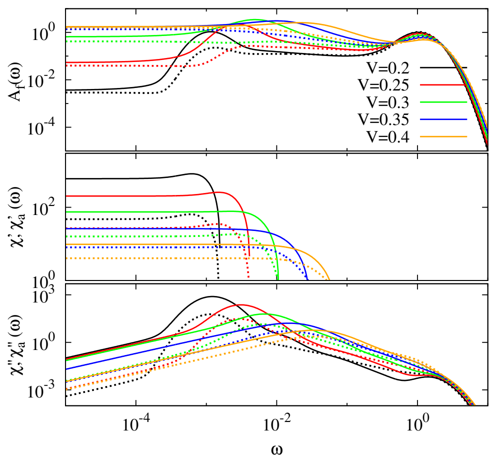

FIG. 2: (color online) Spectral functions (A_f), real and imaginary parts of the spin susceptibilities as functions of the energy for various values of hybridization constant V for impurities sitting on nearest neighbors. The even and odd channels in A_f are represented by solid and dashed lines, respectively. The representations for the spin susceptibilities are the same as in Fig. 1. Here, we choose $U=2$ and $\varepsilon_f = -U/2$. Together with $\Gamma_e(-\omega) = \Gamma_o(\omega)$, the relation $A_{e,o}(\omega) = A_{o,e}(-\omega)$ is satisfied.

We now turn to the second system, i.e, the two impurities are sitting on nearest neighbors. With $\Gamma_{e,o} = \Gamma_0(1 \mp \omega)$, we find that an AFM RKKY interaction is automatically generated $I \approx 0.200\rho_0 J_K^2$, where the Kondo coupling $\rho_0 J_K = 8\Gamma_0/(\pi U) \sim \rho_0 V^2/U$. Because the single-ion Kondo temperature T_K^0 has a different dependence on J_K, we can tune the ratio of I/T_K^0 by tuning J_K with the hybridization constant V (we keep ρ_0 and U fixed constants). In Fig. 2, we show the results of spectral function and the spin susceptibilities. As V increases, I/T_K^0 decreases (see also Fig. 3). Similar to the first system, there are also two distinct phases in two limits: one is the full Kondo resonance state with finite spectral weight at the chemical potential, and another has almost vanishing spectral weight controlled by the inter-impurity spin singlet ground state. However, the sharp transition is replaced by a smooth crossover while the divergence in the staggered spin susceptibility is absent. We can still in general identify two energy scales, a high energy scale where $A_f(\omega)$ and χ'' begin to develop peaks (below $\varepsilon_f + U$), and a low energy scale below which $A(\omega)$ becomes a relative constant and $\chi'' \sim \omega$. The high energy scale has the same origin as T_{coh} in the first system and has the same dependence on I/T_K^0. The low energy scale describes a Fermi liquid temperature T_{FL}, below which Fermi liquid behaviors develop. But it has different behavior compared to T^* in the first system.

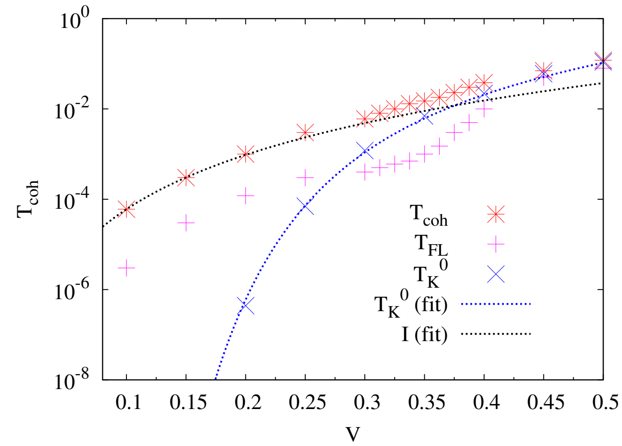

FIG. 3: (color online) The coherence scale as a function of the hybridization constant V. The low energy scale T_{FL} as well as T_K^0 and I are also plotted. T_K^0 is determined from calculations by assuming $\Gamma_{e,o} = \Gamma_0$, and is fitted (in blue dashed line) by $T_K^0 = (0.5/\rho_0)\exp[-(1/\rho_0 J_K) + (1/2)\ln(\rho_0 J_K) + 1.58(\rho_0 J_K)^2]$. The black dashed line denotes the RKKY scale $I = 0.15\rho_0 J_K^2$.

We plot T_{coh} and T_{FL} as functions of V in Fig. 3. Also plotted are the RKKY scale I and the single-impurity Kondo temperature T_K^0 with fitted curves. Here T_K^0 is numerically determined by assuming $\Gamma_{e,o} = \Gamma_0$ (as in the first system) and well fitted by the standard formula. Indeed I and T_K^0 are monotonic functions of V and their ratio I/T_K^0 decreases due to the different dependences on J_K. This phase diagram resembles the Doniach's phase diagram [17] for the heavy fermion lattice as I/T_K^0 is tuned in the same fashion. We find that T_{coh} scales as either I or T_K^0 for $I/T_K^0 \gg 1$ and $I/T_K^0 \ll 1$, respectively. When $I/T_K^0 \sim 1$, T_{coh} is slightly enhanced from T_K^0. This is consistent with the results for the first system. From the analysis on experimental data [1], the heavy fermion materials on the verge of magnetic transitions are suggested to fall in the region $J_K = 0.15$ to 0.20, corresponding to $V = 0.26$ to 0.31 in Fig.3. Indeed, T_{coh} is determined by the RKKY interaction I in this range where $I > T_K^0$. In the two-impurity model, I in this range is bigger than the critical value for the two-impurity quantum critical point and a pseudo-gap state is expected. However, we observe that the spectral function is still finite at the chemical potential with a large T_{FL}, which still resembles a heavy Fermi liquid. This relies on the origin of T_{FL} in this case. Although we assume $\varepsilon_f = -U/2$, the particle-hole symmetry is broken in each even and odd channels. As a result, a potential scattering is present $V_{12}^0(c_{e\sigma}^\dagger c_{e\sigma} - c_{o\sigma}^\dagger c_{o\sigma})$ where

$c_{p\sigma} \sim \sum_{\mathbf{k}} c_{\mathbf{k}\sigma}(e^{i\mathbf{k}\cdot\mathbf{r}_1} \pm e^{i\mathbf{k}\cdot\mathbf{r}_2})$. We also notice that such a parity splitting term can be originated from a hybridization (hopping) term $t_f(f^\dagger_{1\sigma}f_{2\sigma} + h.c.)$ or be generated by a non-parity-splitting potential scattering [11]. In the Bosonization method [9], it generates a relevant interaction $(\psi_f(0) + \psi^\dagger_f(0))(d - d^\dagger)$, where ψ_f is the flavor degree of freedom. This interaction, combining with $(\psi_{sf}(0) - \psi^\dagger_{sf}(0))(d + d^\dagger)$, is responsible for generating Kondo resonance and finite spectral weight at the chemical potential. We denote the Fermi liquid temperature due to this interaction as T_m. Near the quantum critical point, $T^* \sim |I - I_c|$ becomes vanishing while T_m remains finite: the latter determines the Fermi liquid temperature T_{FL}. Its dependence on V in this system is plotted in Fig. 3. There are also other efforts to determine this scale from the particle-hole asymmetry parameters [10, 11]. Between T_{FL} and T_{coh}, there is still an energy range where the intermediate state exists and the system displays non-Fermi liquid behaviors (we also checked this from the self-energy).

A self-consistenly solved two-impurity model can provide a solution to the lattice model within the cluster dynamical mean field theory (CDMFT) approach, where the even and odd parity degrees of freedom correspond to the two momentum points, (0,0,0) and (π,π,π), respectively. From the above results, we would like to make some general arguments on the heavy fermion systems. The RKKY interaction, associated with bare parameters such as V and U, should always be present as an energy scale in the low energy physics. If $I > T^0_K$, the coherence scale is found to be associated with I as an onset of some partial screened Kondo state. Therefore, we expect the lattice coherence scale has the same origin and the same dependence on I. However, it is not directly responsible for the magnetic quantum phase transition, which instead should be reflected in another low energy scale, due to the competition between further Kondo screening and forming magnetic ordered state: either of them can reduce the entropy. The two-energy-scale picture has been indeed found in experiments. For the low energy scale, we would like to bring attention the energy scale T_m, which has been neglected in previous studies on the heavy fermion quantum critical point. It has a different origin than T^* and helps to stabilize the heavy fermi liquid state. In the self-consistency procedure, this scale can vanish due to the Kondo exhaustion when the feedback hybridization functions are not big enough, which is a Mott-type transition. Another possibility is to properly consider the long-wavelength spin fluctuations, a singular form of which can suppress T_m and reveal the criticality associated with T^*. T_m also plays an important role in other systems. For instance, T_m due to the inter-orbital hybridization in two-orbital Hubbard model competes in the formation of the Mott gap.

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